Jammed ~~Glassy~~ Dynamics and Anomalous Diffusion in Self-Assembled Nanoparticle Monolayers

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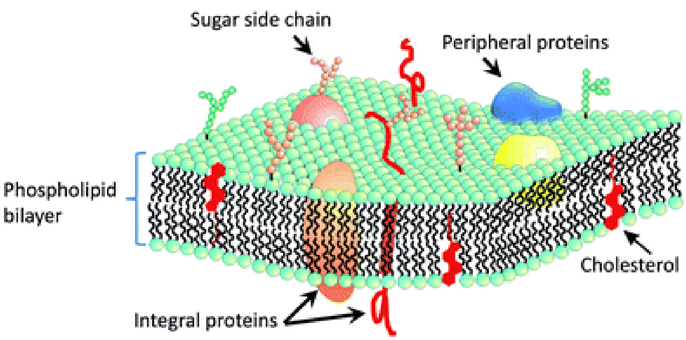
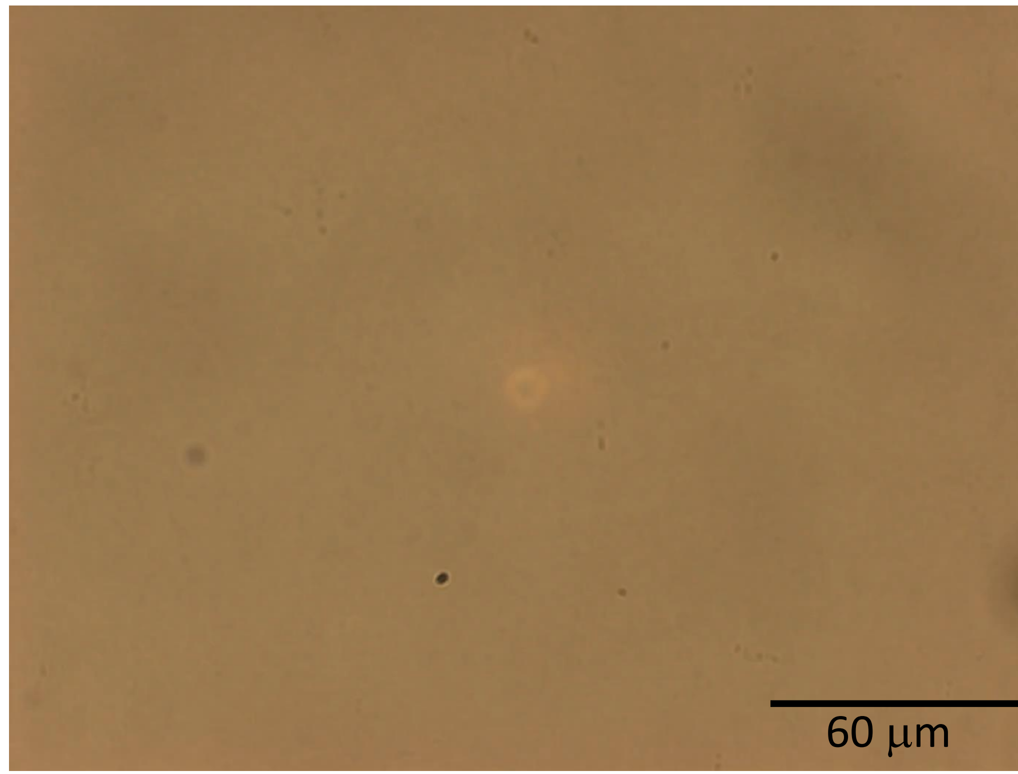
National Science Foundation
WHERE DISCOVERIES BEGIN

Interfacial Structures

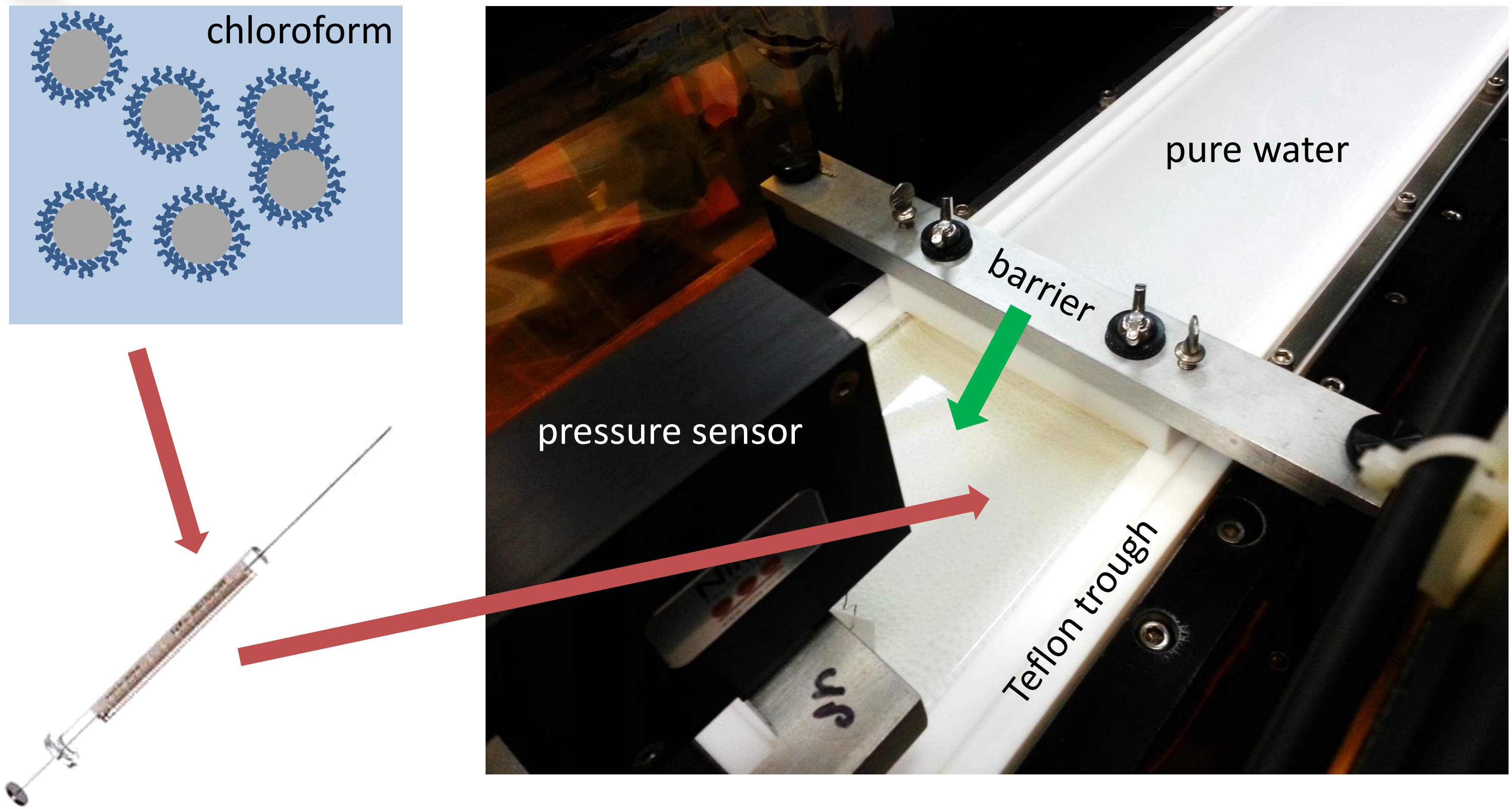


10nm iron oxide nanoparticle film during compression on liquid surface



How do individual particle dynamics affect the film structure?

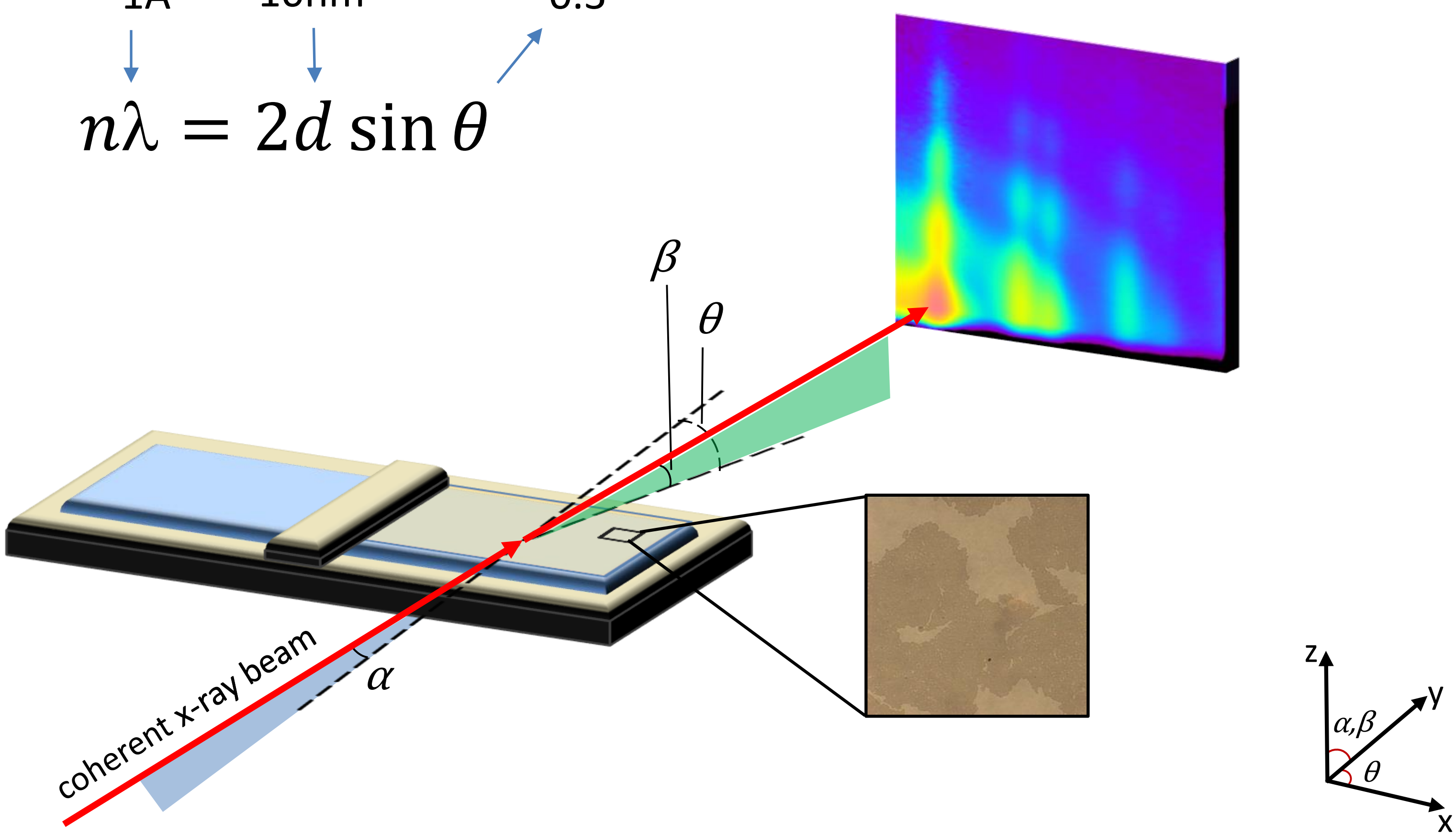
Langmuir-Blodgett Trough



X-Ray Photon Correlation Spectroscopy (XPCS)

~1Å ~10nm ~0.3°

$$n\lambda = 2d\sin\theta$$



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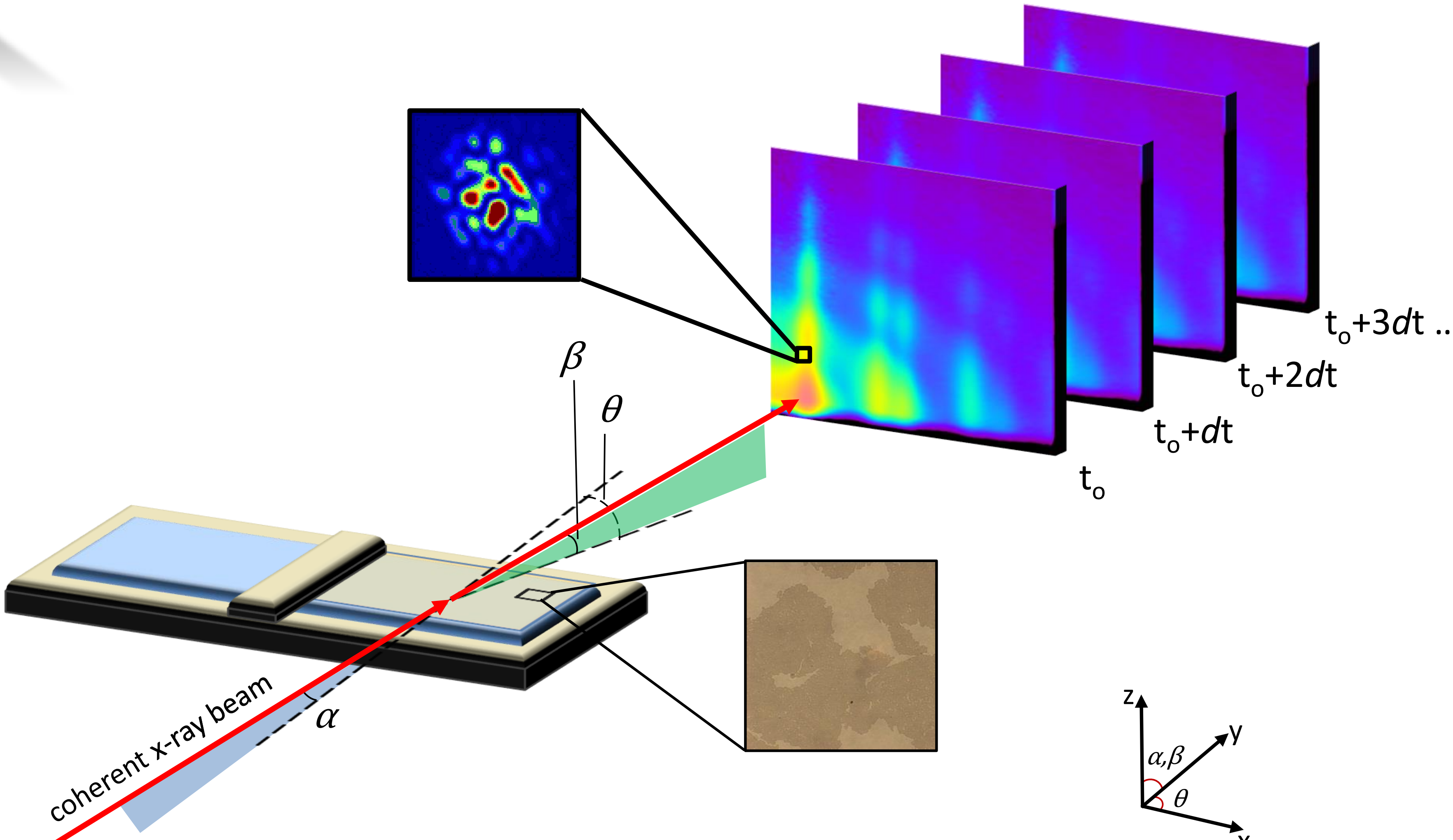
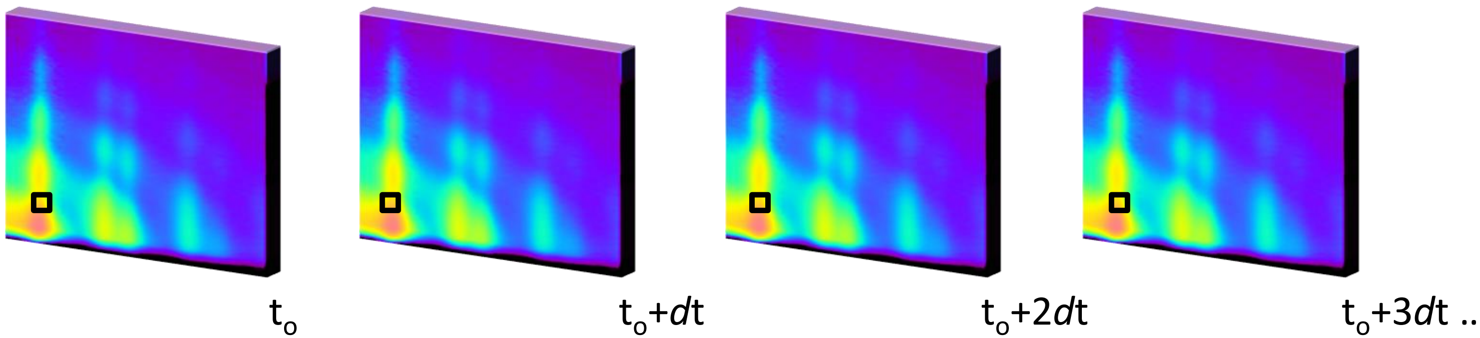
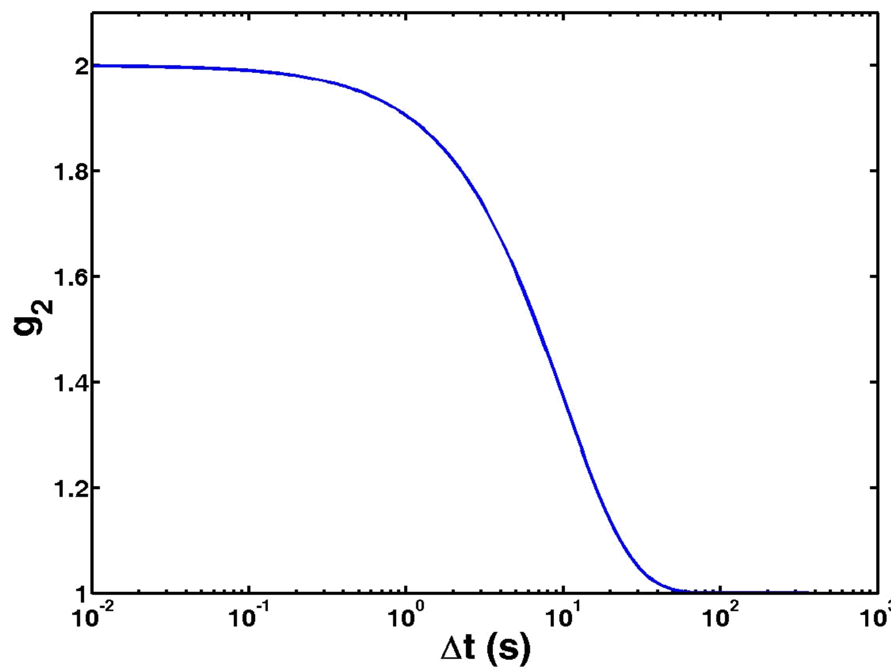


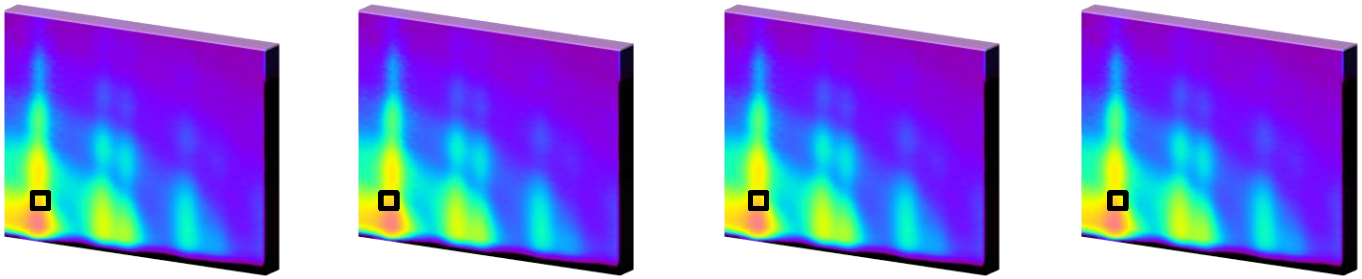
Interparticle Dynamics



$$g_2(\Delta t) = \frac{\langle I(t)I(t+\Delta t)\rangle_t}{{\langle I(t)\rangle_t}^2}$$



Interparticle Dynamics



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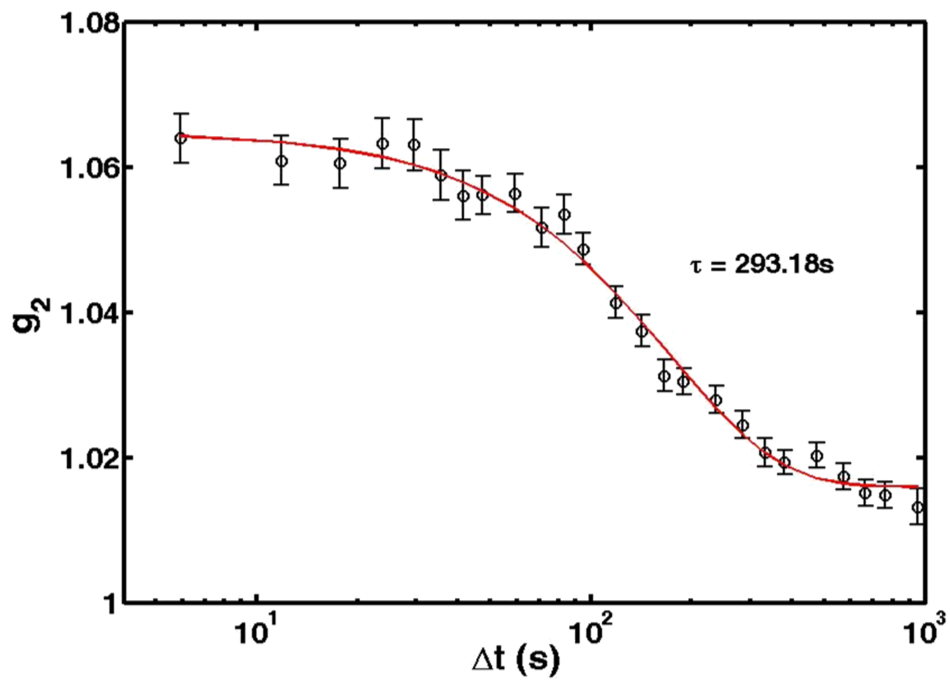


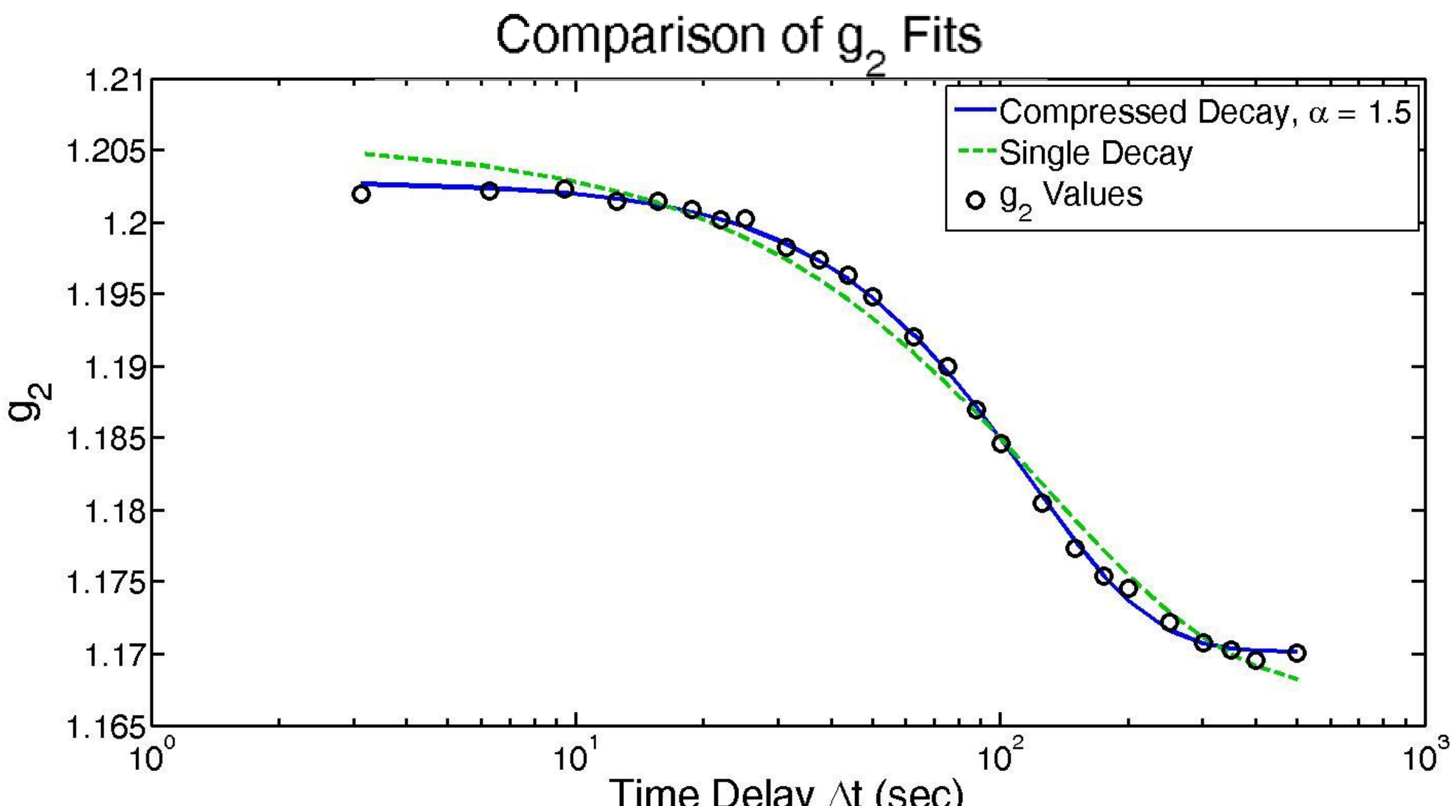
Jamming Transition

Single Exponential (Brownian)

$$g_2 \propto e^{-\frac{t}{\tau}}$$

Compressed Exponential (Jammed State)

$$g_2 \propto e^{-\left(\frac{t}{\tau}\right)^{\alpha}}$$



Cipelletti, L., *et al* (2003). *Faraday Discussions*, *123*, 237–251
Williams, G., & Watts, D.C. (1970). *Transactions of the Faraday Society*, *66*

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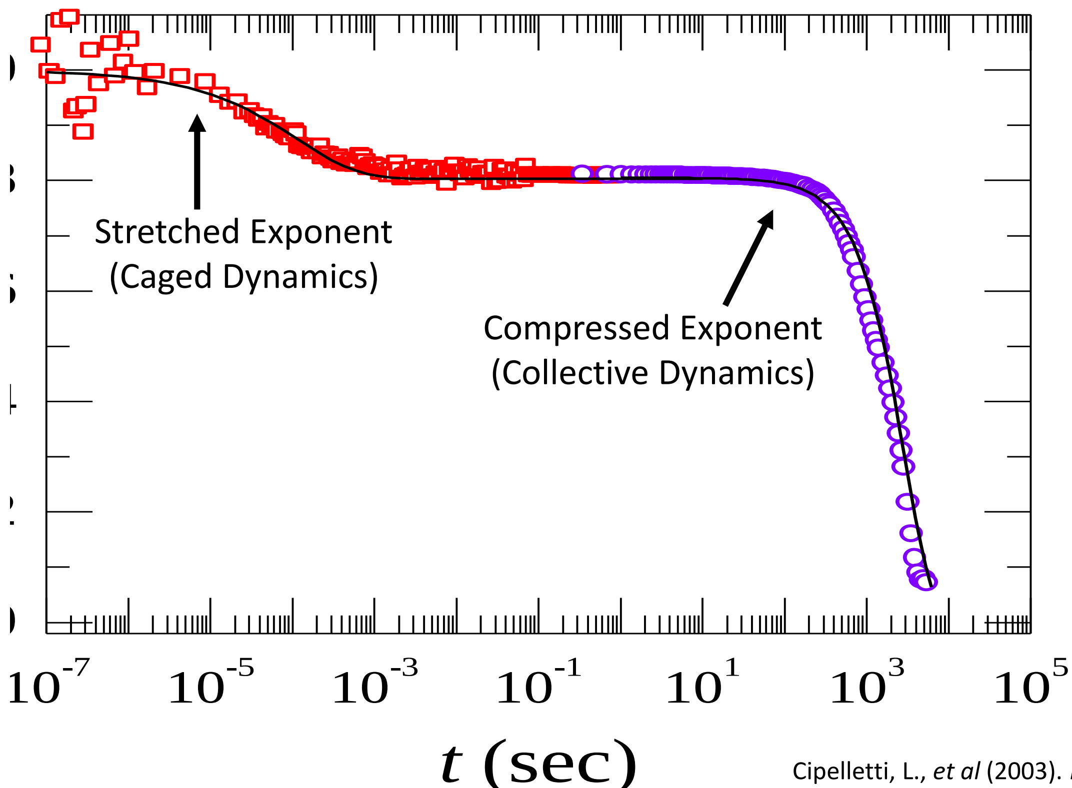


Cipelletti, L., *et al* (2003). *Faraday Discussions*, *123*, 237–251
Williams, G., & Watts, D.C. (1970). *Transactions of the Faraday Society*, *66*

Recurrence of Compressed Exponential

Cipelletti, L., *et. al.* (2003). Universal non-diffusive slow dynamics in aging soft matter. *Faraday Discussions*, *123*, 237–251.

or tens of microns. Remarkably, for all systems the same very peculiar form is found for the final relaxation of the dynamic structure factor: $f(q,t) \sim \exp[-(t/\tau_s)^p]$, with $p \approx 1.5$ and $\tau_s \sim q^{-1}$, thus suggesting the generality of this behavior. Additionally, for all samples the final relaxation slows down with age, although the aging behavior is found to be sample dependent. We propose that the unusual ultraslow dynamics are due to the relaxation of internal stresses, built into the sample at the jamming transition, and present simple scaling arguments that support this hypothesis.

Bouchaud, J.-P., & Pitard, E. (2001). Anomalous dynamical light scattering in soft glassy gels. *The European Physical Journal E*, *6*, 231–236.

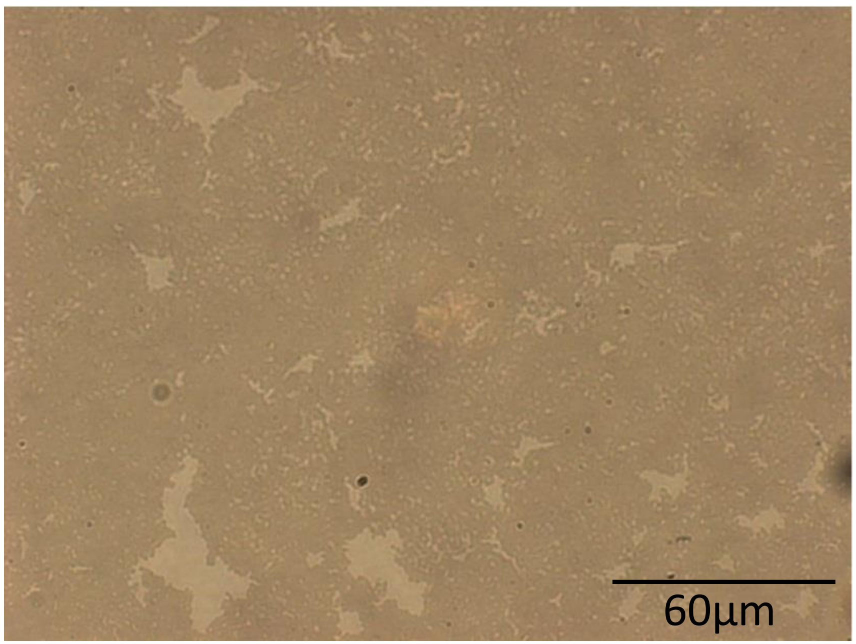
Abstract. We compute the dynamical structure factor $S(q,\tau)$ of an elastic medium where force dipoles appear at random in space and in time, due to "micro-collapses" of the structure. Various regimes are found, depending on the wave vector q and the collapse time θ. In an early-time regime, the logarithm of the structure factor behaves as $(q\tau)^{3/2}$, as predicted in L. Cipelletti, S. Manley, R.C. Ball, D.A. Weitz, Phys. Rev. Lett. **84**, 2275 (2000) using heuristic arguments. However, in an intermediate-time regime we

Bandyopadhyay, R., *et. al.* (2004). Evolution of Particle-Scale Dynamics in an Aging Clay Suspension. *Physical Review Letters*, *93*(22), 228302.

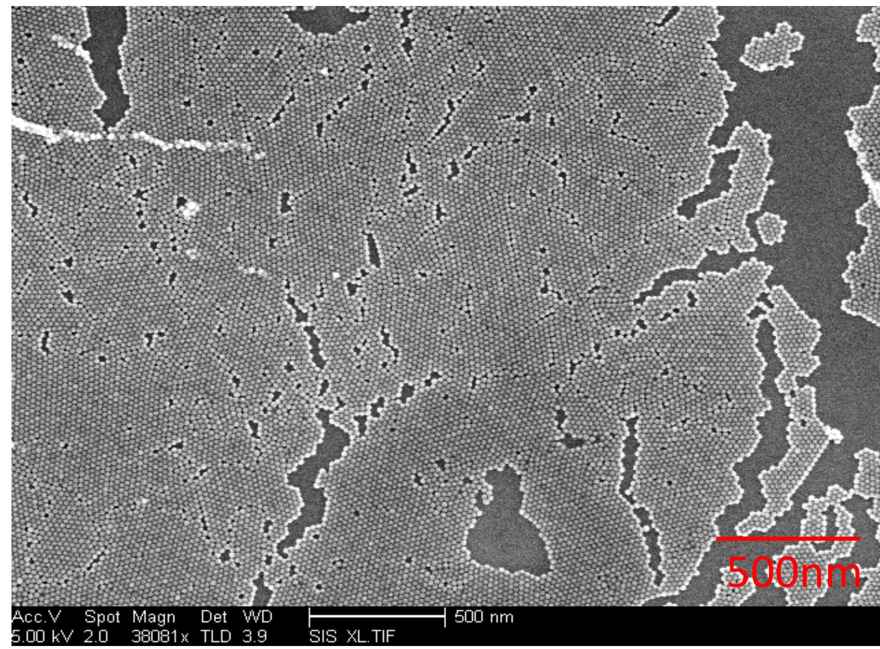
Multispeckle x-ray photon correlation spectroscopy was employed to characterize the slow dynamics of a suspension of highly charged, nanometer-sized disks. At wave vectors q corresponding to interparticle length scales, the dynamic structure factor follows a form $f(q,t) \sim \exp[-(t/\tau)^{\beta}]$, where $\beta \approx 1.5$. The relaxation time τ increases with the sample age t_a approximately as $\tau \sim t_a^{1.8}$ and decreases

Structural Self-Similarity

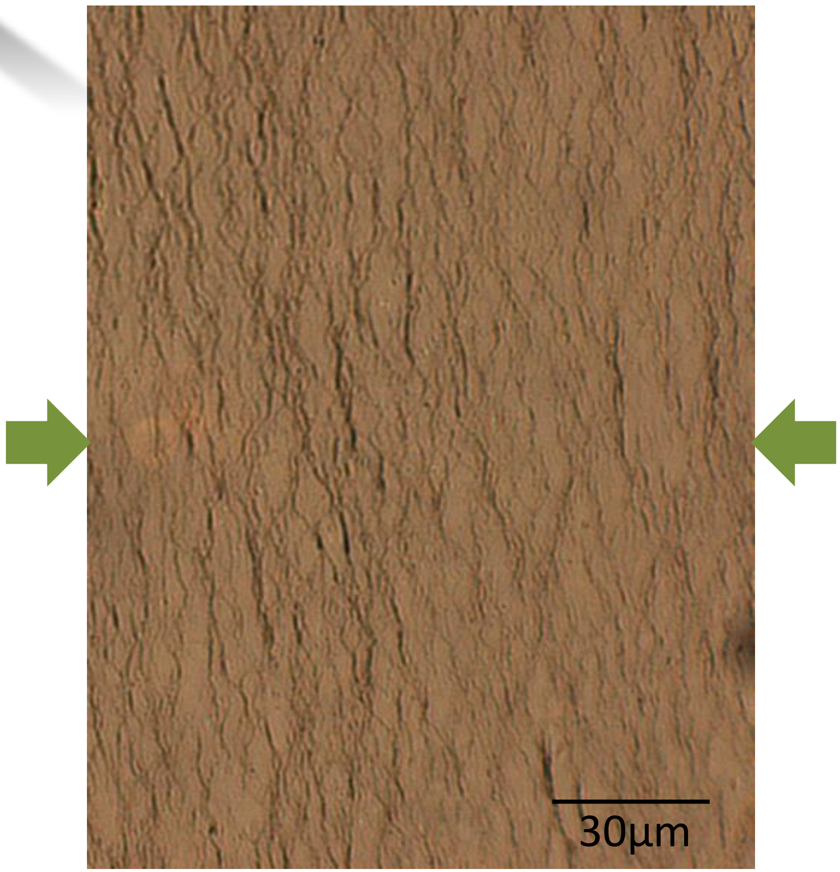
20nm iron oxide particles *in situ* on water surface (optical microscopy)

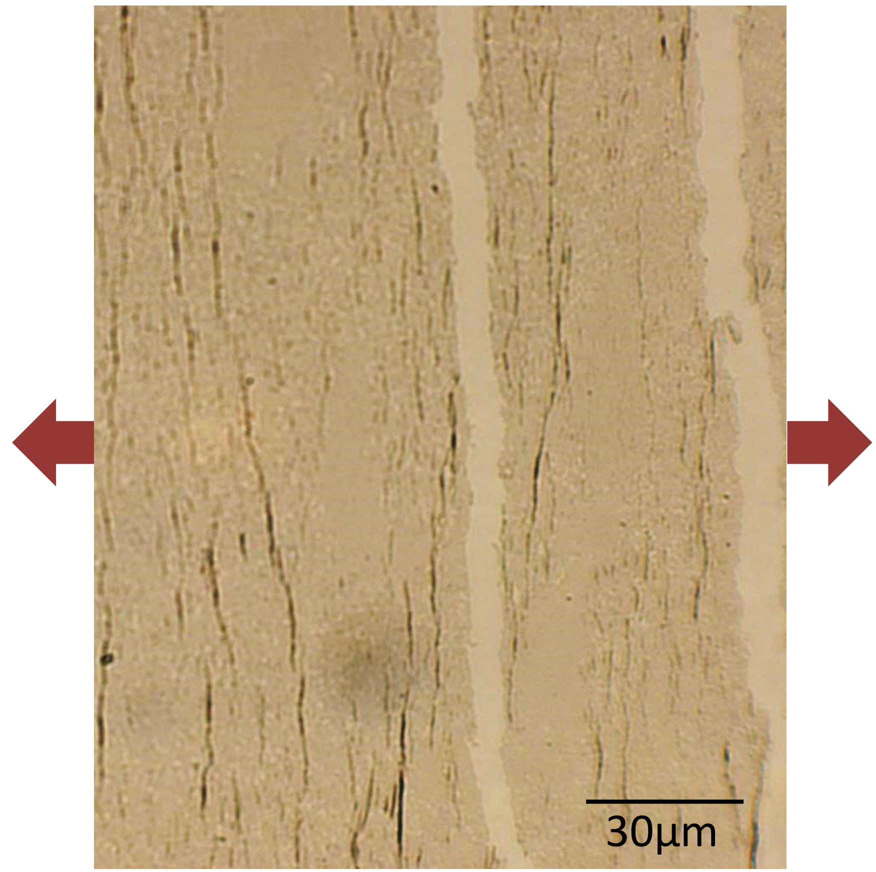


20nm iron oxide particles “stamped” onto silicon substrate (SEM)



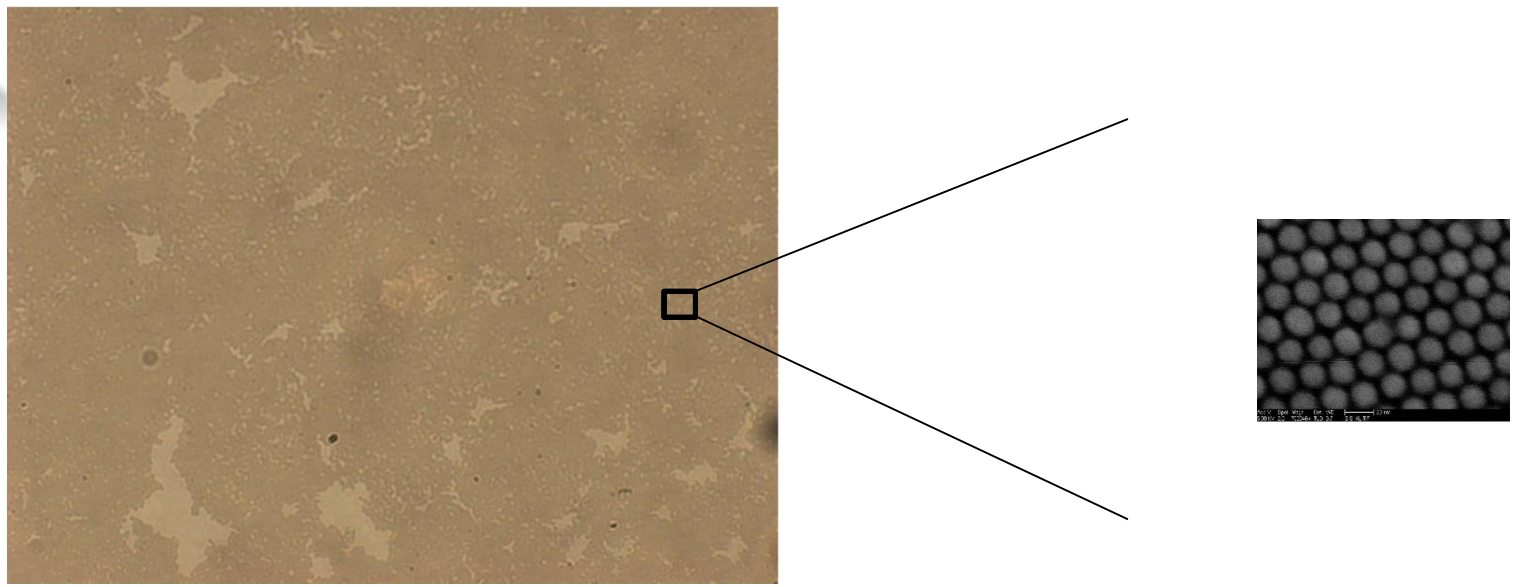
Irreversibility





UCSD

Conclusion

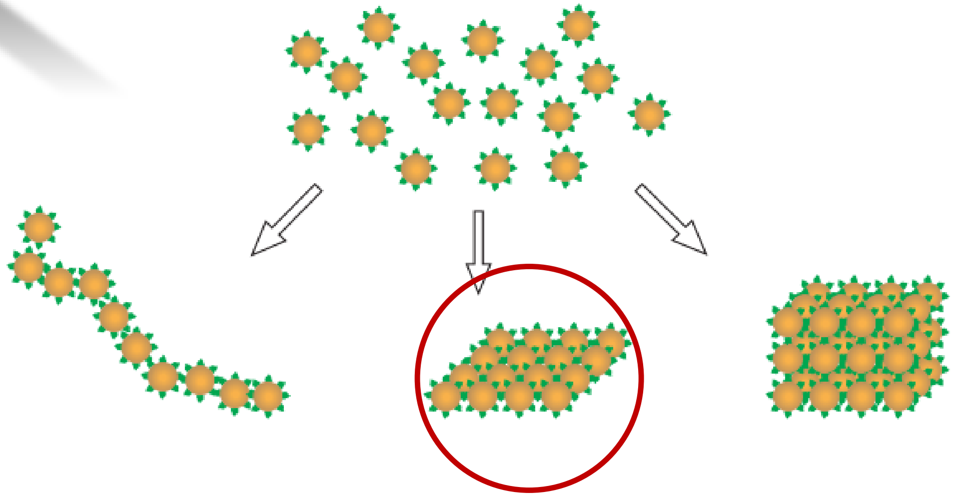


- Quasi-2D system
- Jamming transition
- Compressed 3/2 exponent

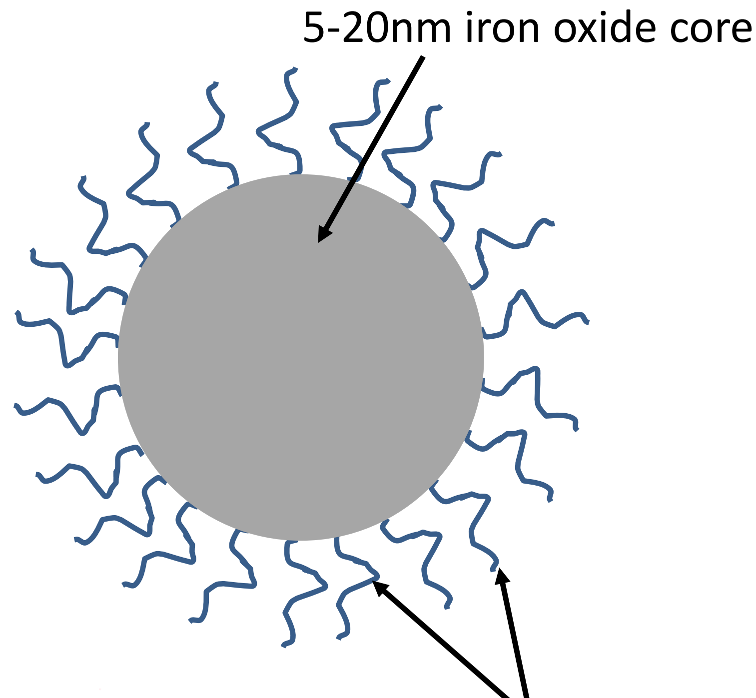
Thank you!

Backup Slides

Liquid Surface Self Assembly

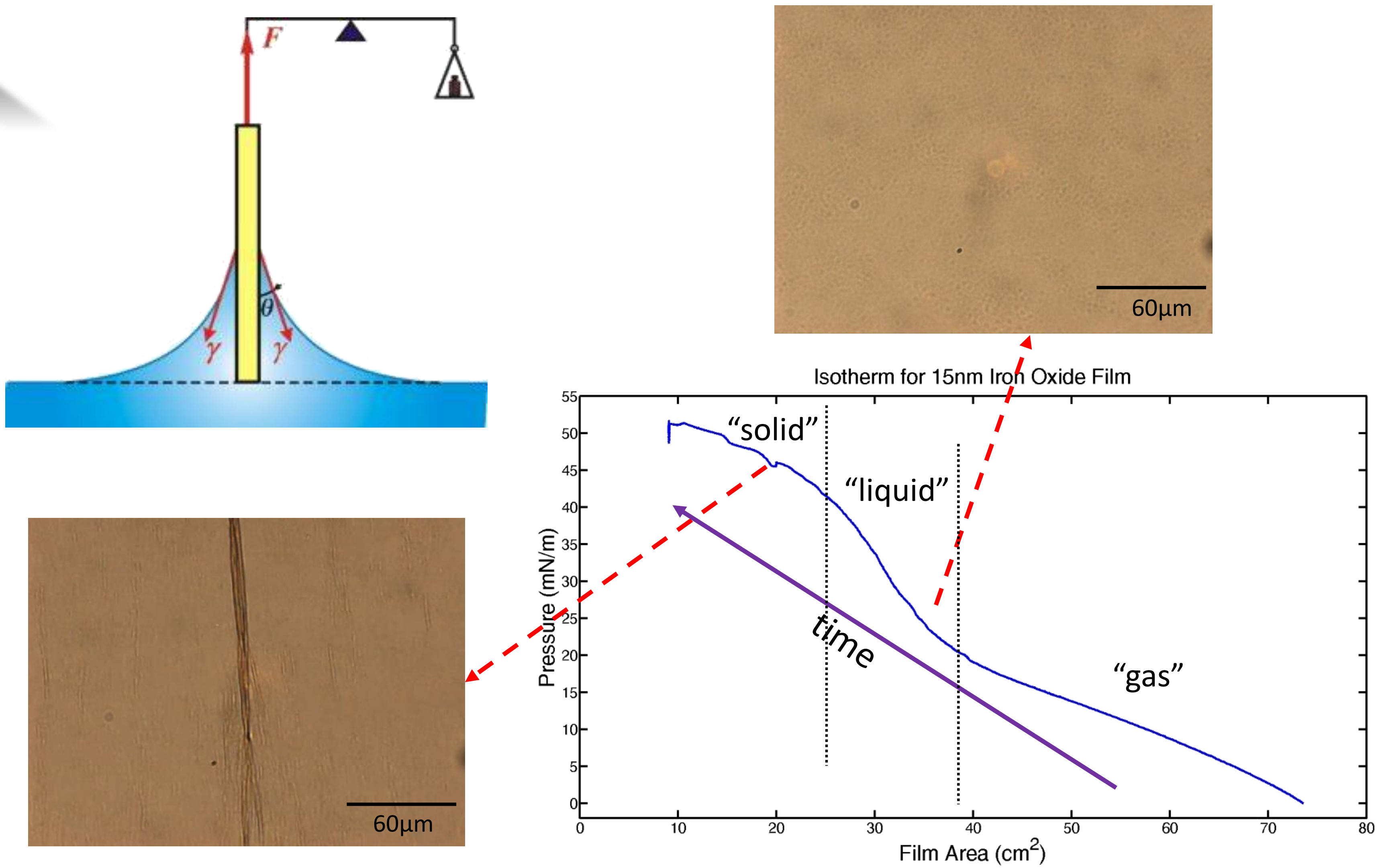


- Van der Waals Force
- Interfacial Forces
- Magnetic Interactions
- Electric Interactions



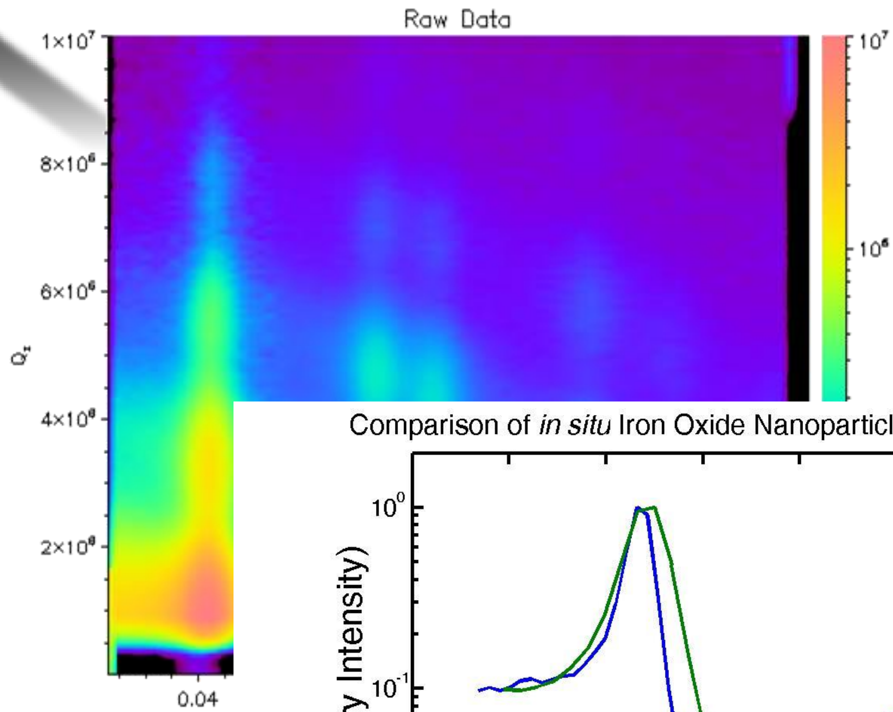
Nie, Z., Petukhova, A., & Kumacheva, E. (2010). *Nature nanotechnology*, *5*(1)

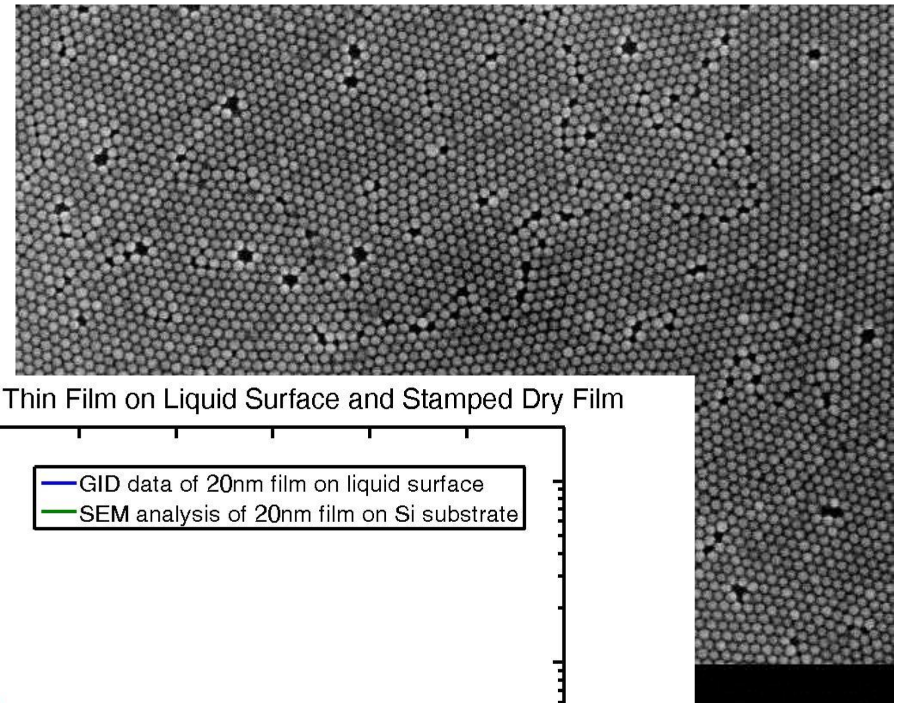
Isotherms

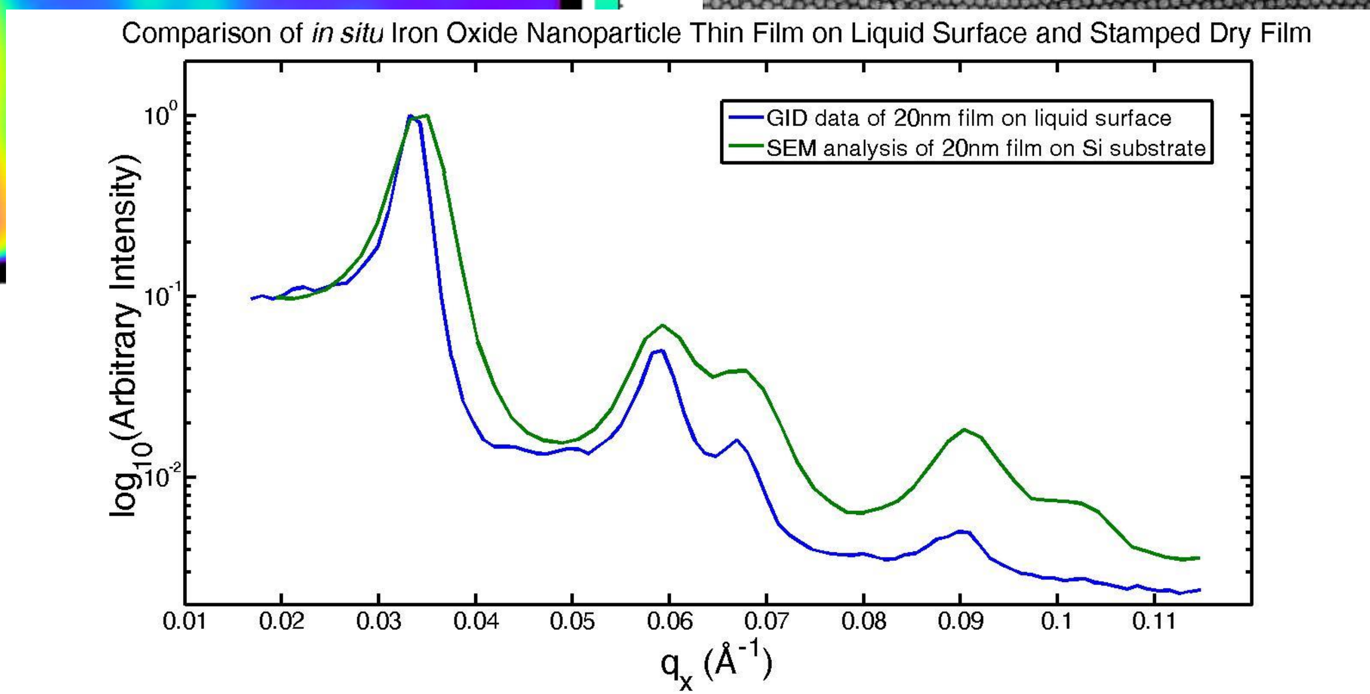


F
θ
γ
γ
60µm
60µm
Isotherm for 15nm Iron Oxide Film
“solid”
“liquid”
“gas”
time
Pressure (mN/m)
Film Area (cm^2)
0 5 10 15 20 25 30 35 40 45 50 55
0 10 20 30 40 50 60 70 80

Preservation of Structure

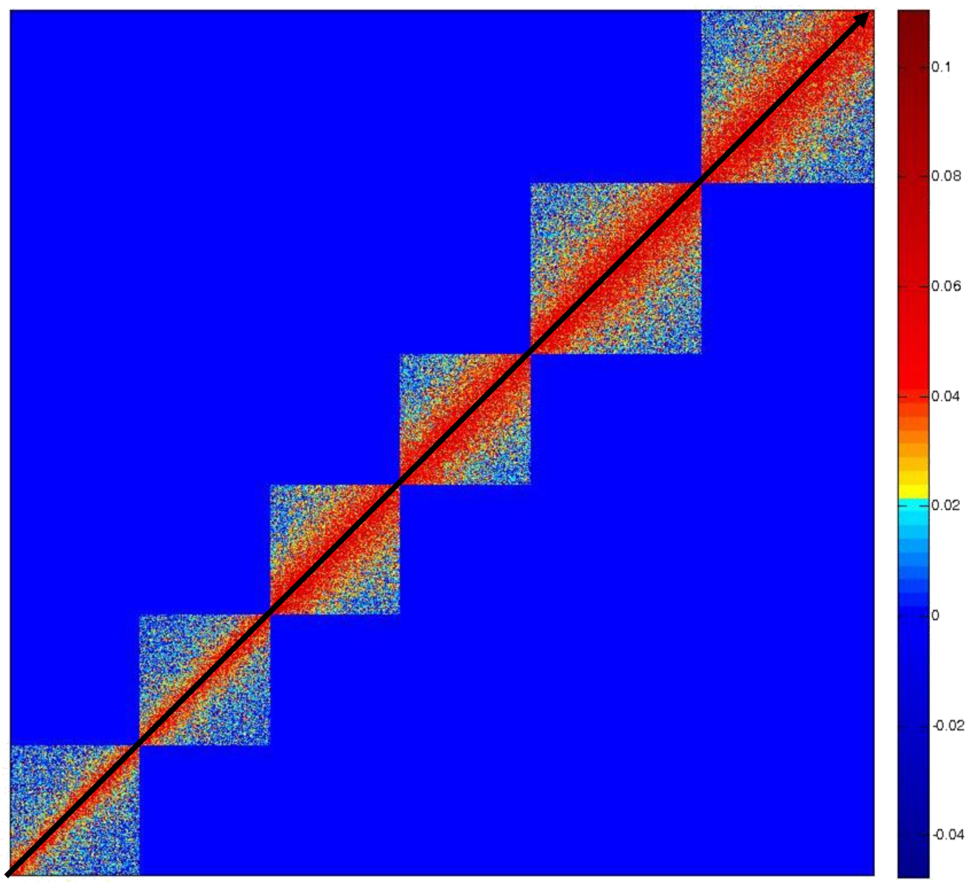


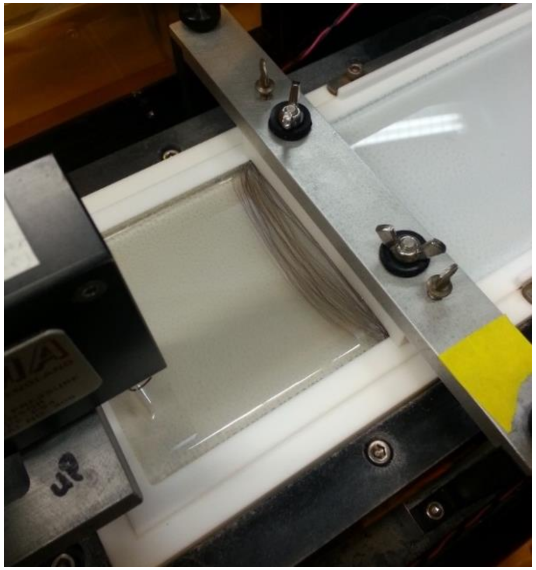


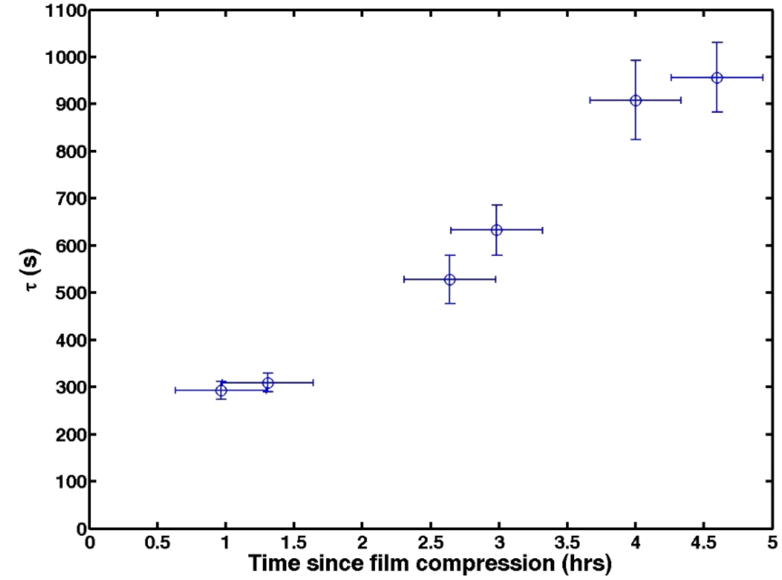


Viscoelasticity

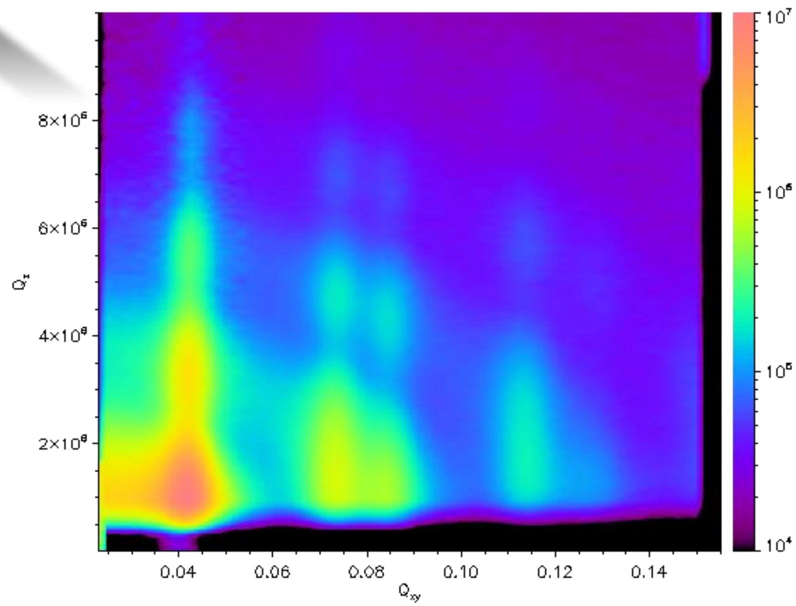
$$g_{2-time}(\Delta t_1, \Delta t_2) = \frac{\langle I(t+\Delta t_1) I(t+\Delta t_2)\rangle_t}{\langle I(t)\rangle^2}$$

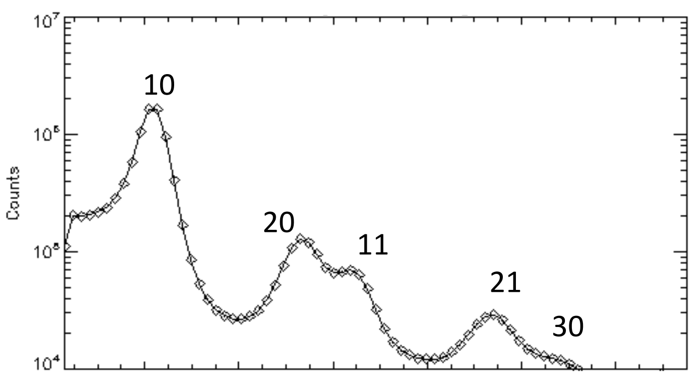


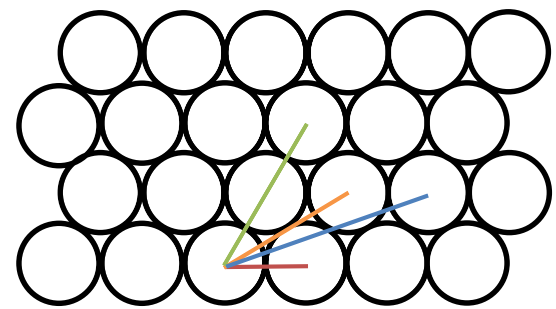




In-Plane Film Structure



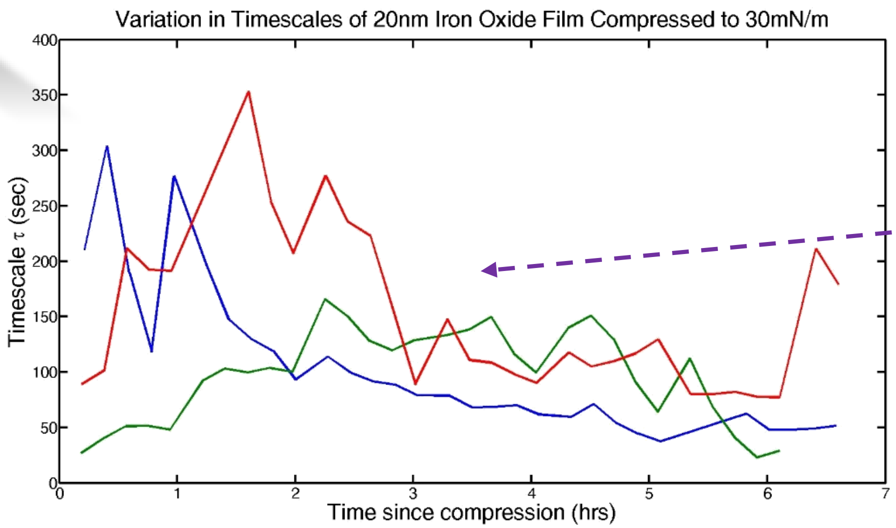




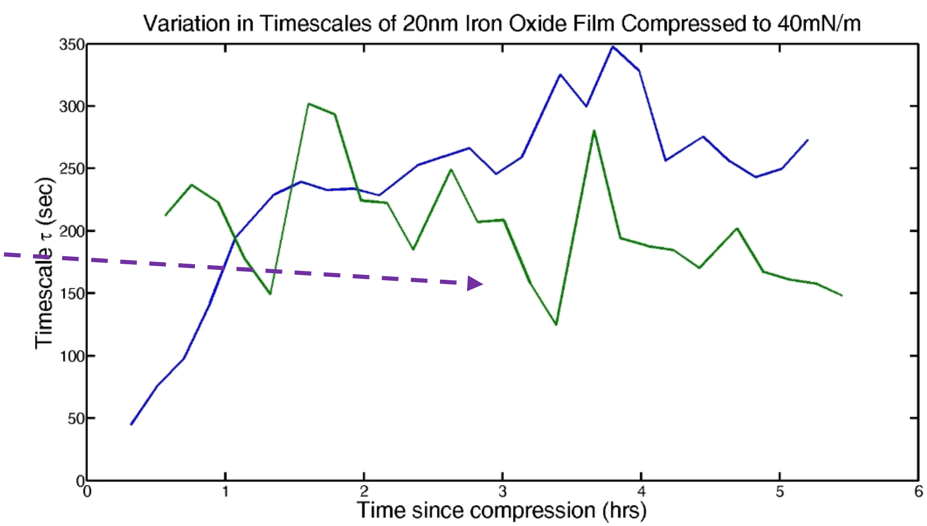
Nearest Neighbor Spacing

	1st	2nd	3rd	4th
Hexagonal Close Packed	1	$\sqrt{3}$ ≈1.73	2	$\sqrt{7}$ ≈2.65
Experiment	1	1.75	2.01	2.74

Timescale-Pressure Dependence



$\bar{\tau}_{30mN/m} = 120s$



$\bar{\tau}_{40mN/m} = 220s$

Q-Dependence of Timescale

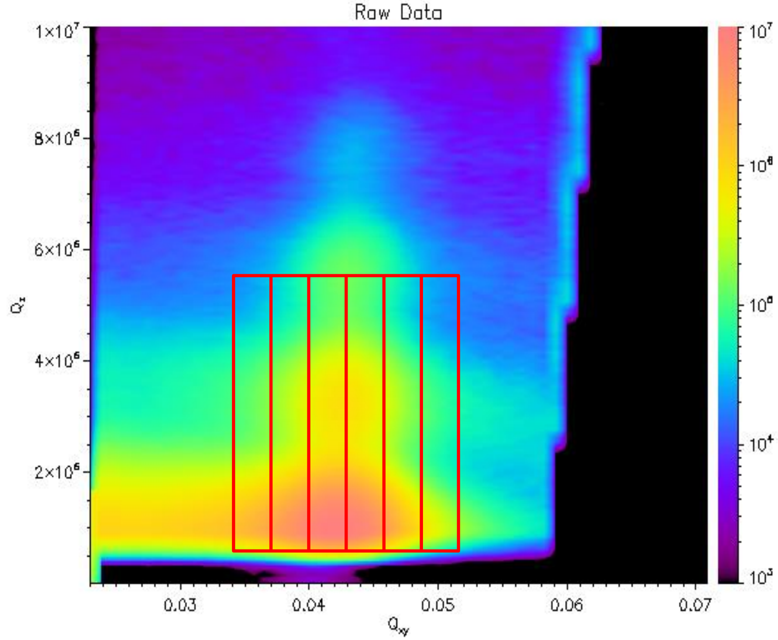
Non-Brownian Motion

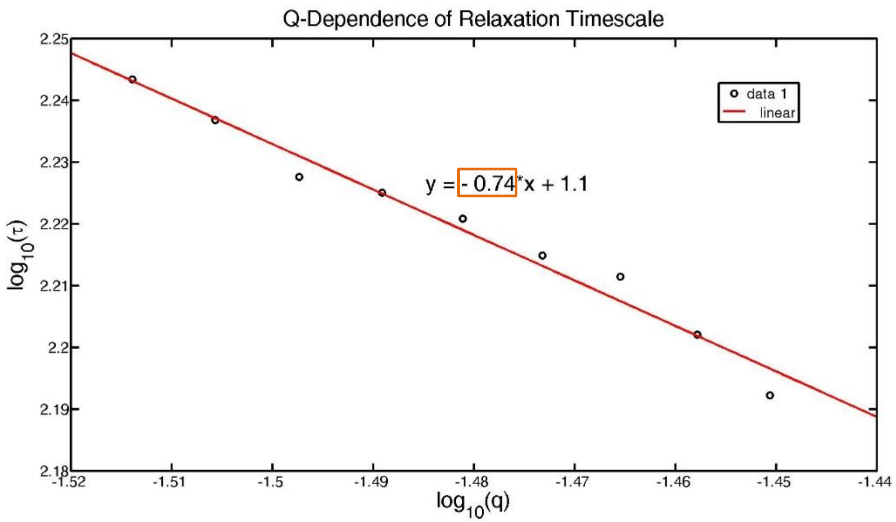
$$\langle x^2 \rangle = 2Dt^{n}$$

$$\langle x^2 \rangle = \left(\frac{2\pi}{q}\right)^2 \qquad t = \tau$$

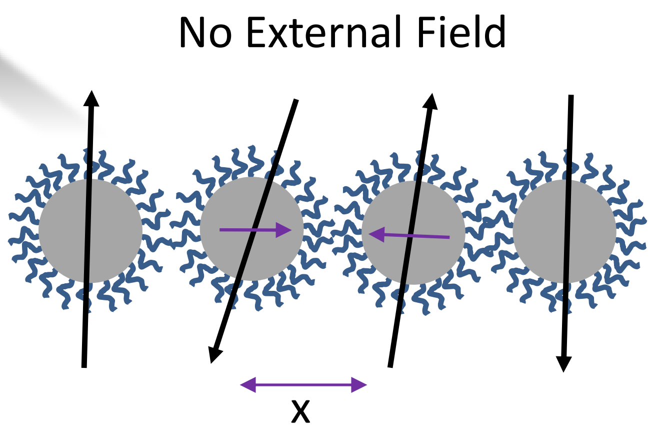
$$\tau = \frac{2\pi^2}{q^2 D}$$

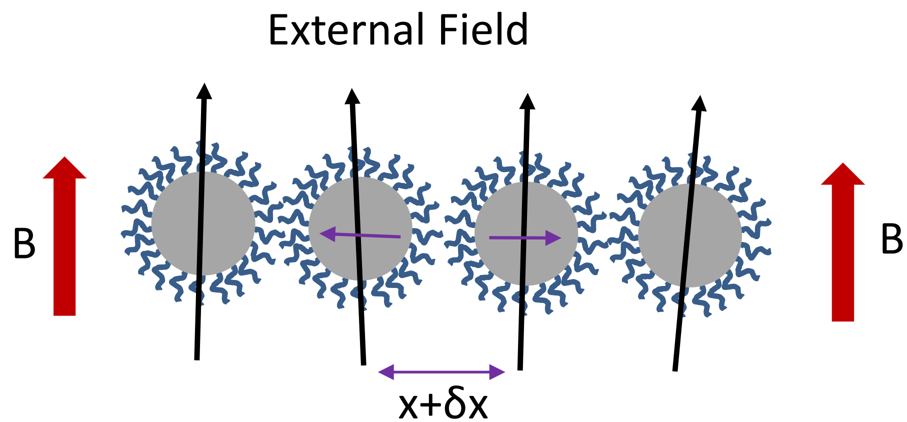
$$\ln \tau = -\frac{2}{n} \ln q + C$$





Magnetic Field Application





Thorek, D.L.J., *et. al.* (2006). *Annals of Biomedical Engineering* , 34(1).